

## \*NOTICES \*

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## CLAIMS

(57)(Claim 6)

[Claim 1]

A polyoxalkylene series polymer (A) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond,

Silicate (B),

an amine compound — and

As a silanol condensation catalyst, they are carboxylic acid tin salt (C) and a non-tin catalyst (E). It is a hardenability constituent to contain,

Silicate (B) is tetra alkoxysilane or its partial hydrolysis condensate,

A carbon atom in which carboxylic acid tin salt (C) adjoins a carbonyl group is the carboxylic acid tin salt which is the 4th class carbon,

A carbon atom in which a non-tin catalyst (E) adjoins a carbonyl group is carboxylic acid which is the 4th class carbon.

A hardenability constituent.

[Claim 2]

A polyoxalkylene series polymer in which a polyoxalkylene series polymer (A) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond introduced an unsaturation group into an end, and a general formula (1):



an allyl group of the carbon numbers 1-20 from which R<sup>1</sup> in a formula and R<sup>2</sup> were the same as or different. When the TETRAORGANO siloxy group shown by any group of the carbon numbers 6-20, an allyl group of the carbon numbers 7-20, or (R<sup>1</sup>)<sub>3</sub>SiO— is shown and R<sup>1</sup> or two or more R<sup>2</sup> exist, they may be the same and may differ. R<sup>1</sup> is a hydrocarbon group of monovalence of the carbon numbers 1-20 here, and three R<sup>2</sup> may be the same and may differ. X shows a hydroxyl group or a hydroxyloxy group, and when two or more X exists, they may be the same and may differ. As for a, 0, 1, 2, or 3b shows 0, 1, or 2, respectively. About b in m bases (SiR<sup>1</sup>)<sub>2</sub>-X<sup>3</sup>-O, they may be the same and may differ. m shows an integer of 0 to 10, however — what satisfies a-sigma b>1 — carrying out — the hardenability constituent according to claim 1, being a polyoxalkylene series polymer obtained by an addition reaction with a hydrosilane compound expressed.

[Claim 3]

The hardenability constituent according to claim 1 or 2 in which a polyoxalkylene series polymer (A) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond is characterized by being a polymer which does not contain an amide segment (—NH—CO—) substantially in a principal chain skeleton.

[Claim 4]

An additive for interior panels containing a hardenability constituent of a description in any 1 paragraph of Claims 1-3, as an object for face panels, or adhesives for car panels.

[Claim 5]

A sealing material for working joint of a building containing a hardenability constituent of a description in any 1 paragraph of Claims 1-3.

[Translation done.]

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## DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention]

This invention relates to the hardenability constituent containing the organic polymer which has a silicon containing functional group (hereinafter, a reactive silicon group) which can construct a bridge by forming a siloxane bond.

[0002]

[Description of the Prior Art]

It is known that the organic polymer which contains at least one reactive silicon group in a molecule thereof is used as a material which constructs a bridge by formation of the siloxane bond accompanied by the hydrolysis reaction of a reactive silicon group, etc., and a rubber-like hardened material is obtained with hygroscopic surface moisture, etc., also in a room temperature.

[0003]

In the polymer which has these reactive silicon groups, a polyoxymethylene series polymer and a polyoxymethylene system polymer are already produced industrially, and are widely used for uses, such as a sealing material, adhesives, and a paint.

[0004]

The adhesives for interior panels, the adhesives for face panels, the adhesives for tiling, the adhesives for stone tensions, when the resin for adhesives used for the adhesives for finishing of wall, the adhesives for car panels, etc., is inferior to stability or creep resistance, an adhesives layer may pass with prudence and the stress from the outside of adherent, it may change by the time, and a panel tile, a stone, etc., may shift. Also in ceiling finishing adhesives or floor finishing adhesives, if inferior to stability or creep resistance, an adhesives layer may pass and it may change by the time, and unevenness of a ceiling surface or a floor line may arise. If the stability of the electrical and electric equipment, an electron, and the adhesives for precision-mechanical-equipment assemblies and creep resistance are bad, an adhesives layer may pass, and it may change by the time, and may be connected with the degradation of apparatus. Therefore, it is called for that the constituent for these adhesives is excellent in stability or creep resistance.

[0005]

The sealing material generally fills up the joined part and crevice between various members, and he is used in order to give watertight and airtightness. Therefore, since the material returns to the use part over a long period of time is very important, excellent in stability or endurance is called for as physical properties of a hardened material. Working joint of a building with an especially large change of joint width (Kasag) the circumference of glass, the circumference of a window frame and a sash, a curtain wall, and various face panels — business — stability and endurance excellent in the constituent used for a sealing material, the sealing material for direct grazing, the sealing material for multiple glass, the sealing material for speed signal generator construction methods, etc., are called for.

[0006]

On the other hand, (the patent documents 1), the (patent documents 2), the (patent documents 3), the (patent documents 4), (The patent documents 5), the (patent documents 6), the (patent documents 7), the (patent documents 8), (The patent documents 9), the (patent documents 10), the (patent documents 11), the (patent documents 12), (The patent documents 13), the (patent documents 14), the (patent documents 15), the (patent documents 16), (The patent documents 17),

[Patent documents 1]  
JP-H10-245482A

[Patent documents 2]  
JP-H10-246484A

[Patent documents 3]  
JP-H10-251552A

[Patent documents 4]  
JP-H10-324793A

[Patent documents 5]  
JP-H10-330630A

[Patent documents 6]  
JP-H10-12473A

[Patent documents 7]  
JP-H11-12480A

[Patent documents 8]  
JP-H11-21463A

[Patent documents 9]  
JP-H11-29713A

[Patent documents 10]  
JP-H11-49869A

[Patent documents 11]  
JP-H11-49970A

[Patent documents 12]  
JP-H11-118631A

[Patent documents 13]  
JP-H11-124509A

[Patent documents 14]  
WO No. 47839 [ 88 to ]

[Patent documents 15]  
JP 2000-34391A

[Patent documents 16]  
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they may be the same and may differ, R' is a hydrocarbon group of monovalence of the carbon numbers 1-20 here, and three R may be the same and may differ. X shows a hydroxyl group or a hydroxylic basis, and when two or more X exists, they may be the same and may differ. As for a, 0, 1, 2, or 3b shows 0, 1, or 2, respectively. About b in m base (SiR<sub>2</sub>-2-X<sub>2</sub>O) may be the same and may differ, m shows an integer of 0 to 19, however — what satisfies a+3m b=1 — carrying out — it is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using a hardenability constituent which is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed.

An organic polymer in which an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment introduced an unsaturation group into an end, and general formula (2) :



X in a formula shows a hydroxyl group or a hydroxylic basis, and three X may be the same and it may differ. It is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using a hardenability constituent which is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed.

[0048] The 4th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond in said invention, and this organic polymer is a general formula (3) :

$$\text{O-R}^2\text{-CH(OR}^1\text{)-CH}_2\text{-(SR}^1\text{-2-X}_2\text{O)}_m\text{-SR}^2\text{-2-X}_2\text{O} \quad (3)$$

(R<sup>1</sup> formula a divalent organic group of the carbon numbers 1-20 which contain one or more sorts chosen from a group which consists of hydrogen, oxygen, and nitrogen as a composition set) [ show and ] R<sup>1</sup>, R<sup>2</sup>, X, a, b, and m — the above — it is the same — it is related with stability, endurance, and creep resistance corrective strategy of a hardened material using a hardenability constituent, which is an organic polymer (A3) which has a structure part with which it is expressed.

[0049] An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (4) :

$$\text{O-R}^2\text{-C(OR}^1\text{)-CH}_2 \quad (4)$$

An organic polymer which introduced an unsaturation group expressed with (R<sup>2</sup>) is the same as the above, and general formula (1) :

$$\text{H-SiR}^2\text{-2-X}_2\text{O)}_m\text{-SiR}^2\text{-2-X}_2\text{O} \quad (1)$$

It is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in the above using a hardenability constituent which is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed with (R<sup>1</sup> in formula, R<sup>2</sup>, X, a, b, and m are the same as the above).

[0050] An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (5) :

$$\text{O-R}^2\text{-CH(OR}^1\text{)-CH}_2\text{-SiX}_3 \quad (5)$$

(R<sup>1</sup> formula and X are the same as the above) — it is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using a hardenability constituent which is an organic polymer which has a structure part with which it is expressed.

[0051] An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment, it is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using a principal chain skeleton a hardenability constituent which is an organic polymer which does not contain an amide segment (—NH—CO—) substantially.

[0052]

A silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (6) :



(three R<sup>1</sup> is an organic group of monovalence of the carbon numbers 2-20 independently among a formula, respectively) — it is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using a hardenability constituent which is a basis expressed.

[0053] It is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either, wherein a silicon containing functional group which can construct a bridge by forming a siloxane bond uses a hardenability constituent which is a triethoxy silyl group as a desirable embodiment.

[0054]

The 5th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond is related with this layer hardenability corrective strategy using an silicon an organic polymer (A1) which is a silicon containing functional group which consists of three or more hydroxylic bases, and a hardenability constituent containing an organic tin catalyst

[0055]

The 6th is an organic polymer (A1) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. Adhesives for interior panels containing silicate (B). Adhesives for floor panels, adhesives for tiling, adhesives for stone tensions, ceiling finishing (B). Adhesives for face panels, adhesives for floor panels, adhesives for finishing of wall,

adhesives for car panels, the electrical and electric equipment, an electron and adhesives for precision-mechanical-equipment assemblies, a sealing material for direct grouting, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0056]

The 7th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond, it is characterized by containing an organic polymer (A1) which is a silicon containing functional group which has three or more hydroxylic bases on silicon. Adhesives for interior panels, adhesives for floor panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, it is related with the electrical and electric equipment, an electron and adhesives for precision-mechanical-equipment assemblies, a sealing material for direct grouting, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0057]

A main chain of an organic polymer (A1) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment. Using a hardenability constituent which is an acrylic ester system copolymer manufactured by a living radical-polymerization method (meta) to the above by which it is characterized A description. Adhesives for interior panels, adhesives for floor panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, it is related with the electrical and electric equipment, an electron and adhesives for precision-mechanical-equipment assemblies, a sealing material for direct grouting, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0058]

As a desirable embodiment, containing silicate (B) further to said either by which it is characterized A description. Adhesives for interior panels, adhesives for floor panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, it is related with the electrical and electric equipment, an electron and

adhesives for precision-mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0059]

As a desirable embodiment, containing carboxylic acid an salt (O) further to said either by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0060]

As a desirable embodiment, containing an organic tin catalyst (D) further to said either by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0061]

The 8th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of the invention. It is characterized by this organic polymer being an organic polymer (A2) which averages per molecule a silicon containing functional group which can construct a bridge by forming a siloxane bond and has it 1.7-5 pieces. Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0062]

A silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment, That it is a silicon containing functional group which has three or more hydroxylic bases on the above by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0063]

An organic polymer in which an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment introduced an unsaturation group into an end and a general formula (1):



That it is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed with (R<sup>1</sup> in terms of R<sup>2</sup>, X, a, b, and m are the same as the above) to said either by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0064]

An organic polymer in which an organic polymer which has a silicon containing functional group which

can construct a bridge by forming a siloxane bond as a desirable embodiment introduced an

unsaturation group into an end and a general formula (1):



That it is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed

with (R<sup>1</sup> in terms of R<sup>2</sup>, X, a, b, and m are the same as the above) to said either by which it is

characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for

tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for

can construct a bridge by forming a siloxane bond as a desirable embodiment introduced an

unsaturation group into an end and a general formula (2):



That it is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed

with (R<sup>1</sup> in terms of R<sup>2</sup>, X, a, b, and m are the same as the above) to said either by which it is

characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for

stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall,

adhesives for car panels, It is related with the electrical and electric equipment, an electron and

mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple

glass, a sealing material for speed signal generator construction methods, or a sealing material for

working joint of a building.

[0065]

The 9th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of the invention, and this organic polymer is a general formula (3):



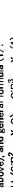
It is characterized by being an organic polymer (A3) which has a structure part expressed with (R<sup>1</sup> in terms of R<sup>2</sup>, R<sup>3</sup>, X, a, b, and m are the same as the above), Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0066]

An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (4):



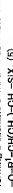
An organic polymer which introduced an unsaturation group expressed with (R<sup>1</sup> is the same as the above), and general formula (1):



That it is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed with (R<sup>1</sup> in terms of R<sup>2</sup>, X, a, b, and m are the same as the above) to the above by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0067]

An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (5):



(R<sup>1</sup> in terms of X are the same as the above) — a description to said either being an organic polymer which has a structure part with which it is expressed, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0068]

An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment, That it is an organic polymer which does not





dichloroacetate and xylene dicarboxylate isophthalate dicarboxylate. What is obtained from a reaction of polyisocyanate compounds, such as various isocyanate system polyisocyanate compounds, such as hexamethylene diisocyanate, and polyol which has various kinds of above-mentioned principal chain skeletons can be a polyurethane compound.

[0096] If there are many amide segments (—NH—CO—) generated in a principal chain skeleton based on said urethane bond, viscosity of an organic polymer will become high and will serve as a bad constituent of workability. Therefore, as for quantity of an amide segment occupied in a principal chain skeleton of an organic polymer, it is preferred that it is 3 or less of the weight, it is more preferred that it is 1 or less of the weight, and it is most preferred that an amide segment is not included substantially.

[0097] A reactive silicon group contained in an organic polymer which has a reactive silicon group is a basis which can construct a bridge by forming a siloxane bond by the reaction which has a hydroxyl group or a hydrolytic basis combined with a silicon atom, and is accelerated by a silanol condensation catalyst. As a reactive silicon group, it is a general formula (11):



in which group of the carbon numbers 1-20 from which  $R^1$  in formula and  $R^2$  were the same or different. When the Tert ORGANO siloxy group shown by aryl group of the carbon numbers 6-20, an aryl group of the carbon numbers 1-20, or (R<sup>3</sup>)<sub>2</sub>SiO<sup>-</sup> is shown and R<sup>1</sup> or two or more R<sup>2</sup> exist, they may be the same and may differ. R<sup>1</sup> is a hydrocarbon group of monovalence of the carbon numbers 1-20 here, and three R<sup>2</sup> may be the same and may differ. X shows a hydroxyl group or a hydrolytic basis, and when two or more X exists, they may be the same and may differ. As for a, 1, 2, or 3b shows 0, 1, or 2, respectively. About X in bases (SiR<sup>1</sup>)<sub>2</sub>-X<sub>a</sub>-O<sup>-</sup>, they may be the same and may differ, m shows an integer of 0 to 19, however — what satisfies a+b=1 — carrying out — a basis expressed is added.

[0098] It is not limited but what is necessary is just a conventionally publicly known hydrolytic basis especially as a hydrolytic basis. Specifically, a hydrogen atom, a halogen atom, an alkoxy group, an acyloxy group, a KETOKISHI mate group, an amino group, an amide group, an acid-amide group, an aminoxy group, a sulfinyloxy group, an alkenyloxy group, etc. are mentioned, for example. Among these, a hydrogen atom, an alkoxy group, an acyloxy group, a KETOKISHI mate group, an amino group, an amide group, an aminoxy group, a sulfinyloxy group, and an alkenyloxy group are preferred. Hydrolysis nature is quiet and a viewpoint of handling or a conc to especially an alkoxy group is preferred.

[0099] A hydrolytic basis and a hydroxyl group can be combined with two silicon atom in the 1-3 ranges, and (R<sup>1</sup> groups) have 1-3 preferred ranges. When a hydrolytic basis and a hydroxyl group join together in a range of more than 3 a reactive silicon group, they may be the same and may differ.

In particular, it is a general formula (12):



(R<sup>2</sup> and X are the same as the above among a formula.) Since a reactive silicon group expressed with an integer of 1-3 is easy to resolve, a is preferred.

[0101]

As an example of R<sup>1</sup> in the above-mentioned general formula (11) and (12), and R<sup>2</sup>. For example, aryl groups, such as aryl groups, such as cycloalkyl groups, such as silyl groups, such as a methyl group, and an ethyl group, and a cyclohexyl group, and a phenyl group, and benzyl, the Tert ORGANO siloxy group R<sup>1</sup> is indicated to be by —SiO<sup>-</sup>— which is a methyl group, a phenyl group, etc. (R<sup>2</sup>, etc. are methyl. Especially in these, a methyl group is preferred.

[0102]

As more concrete illustration of a reactive silicon group, a trimethoxysilyl group, a triethoxysilyl group, a triisopropoxysilyl group, a dimethoxymethyl silyl group, a dichloromethylsilyl group, and a

dichloropropoxysilyl group are mentioned.

[0103]

Especially in this invention, an organic polymer which has the silicon containing functional group (that is, number of a term of a general formula (11) is three or more) which three or more hydrolytic bases combined on silicon in an organic polymer of the (A) ingredient can be used as an ingredient.

[0104]

A hardened material which three or more hydrolytic bases had combined this (A1) ingredient on silicon, and constructed the bridge by a silanol condensation reaction of that reactive silicon group. Good stability is shown and remarkable creep resistance and an endurance improvement effect are shown as compared with a case of reactive silicon group containing organic polymer which has two or less hydrolytic bases.

[0105]

(A1) As for the number of a term of a general formula (11) of an ingredient, it is more preferred that it is 3-5, and especially 3 is preferred. Also in it, since [ that its improvement effect of the stability of a hardenable constituent of this invention, endurance, and creep resistance is especially large and ] the Tert alkoxy silyl groups has the good availability of a raw material, it is preferred. Thing of an alkoxy group of the carbon numbers 1-20 is preferred, the thing of the carbon numbers 1-10 is more preferred, and its thing of the carbon numbers 1-4 is still more preferred here. Specifically, a trimethoxysilyl group and a triethoxysilyl group are the most preferred. Hardenable may become a hardened material when a carbon number is larger than 20.

[0106]

Generally, if weight of reactive silicon group containing organic polymer in a hardenable constituent becomes low, it is known that the endurance of a hardened material obtained will fall to low. However, if an ingredient (A1) of this invention is used as a reactive silicon group containing organic polymer, high endurance is maintainable even if it makes low weight % of reactive silicon group containing organic polymer in a hardenable constituent. Therefore, five to 28% of the weight, when it is 15 to 24 % of the weight especially preferably, since a rate of an ingredient (A1) in a hardenable constituent is compatible in low cost and high endurance, it is more preferably preferred [ rate ] ten to 26% of the weight.

[0107]

Especially in this invention, an organic polymer which has the Tert alkoxy silyl groups of the carbon numbers 2-20 can be used as a (A4) ingredient in an organic polymer of an ingredient (A1). Namely, general formula (6):



(three R<sup>4</sup> is an organic group of monovalence of the carbon numbers 2-20 independently among a formula, respectively) — an organic polymer which has a basis expressed can be used as a (A4) ingredient.

[0108]

It is known that methanol generated in connection with a hydrolysis reaction of a methoxy silyl group has peculiar toxicity of causing an obstacle of an optic nerve. On the other hand, since a carbon number of an alkoxy group which combines the (A4) ingredient with a silicon atom is 2 to 20, toxic high methanol is not contained in alcohol generated in connection with a hydrolysis reaction of a reactive silicon group, but serves as a constituent with high safety at it.

[0109]

(A4) It is preferred that it is especially 2-4, and when it is 2, it serves as ethanol, and since alcohol generated by hydrolysis has the highest safety, it is the most preferred [ as for a carbon number of R<sup>4</sup> of a general formula (6) of an ingredient, it is more preferred that it is 2-10, and / or alcohol ].

Specifically, a triethoxy silyl group is the most preferred. When a carbon number is larger than 20, while the hardenable of a hardenable constituent may become late, an anesthetic action and stimulation of alcohol to generate may be large.

[0110]

Especially in this invention, a principal chain skeleton can use as a (A5) ingredient what is polyoxymethylene in an organic polymer of the (A4) ingredient. Namely, general formula (6):





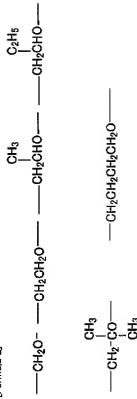
handling 1, stability 1 of a hardenability constituent obtained 1, endurance, and creep resistance \*\* to 1, dimethoxysilane is the most preferred.

[0127] Although a method of introducing into an unsaturation binding site of an organic polymer a reactive silicon group and a reactive silicon group as a synthetic method by a radical addition reaction under a radical initiator and/or radical source-to-release existence, for example, etc. are mentioned, it is not limited in particular. As an example of a compound of having said silylalkyl group and a reactive silicon group. For example, although gamma-mercaptopropyltrimethoxysilane, gamma-mercaptopropylmethyl dimethoxysilane, gamma-mercaptopropylmethyl diethoxysilane, etc. are raised, it is not limited to these.

[0128] As mentioned above, as for a silane compound which three hydroxylic bases have combined with one silicon atom, such as trimethoxysilane, disopropoxysilane may advance. As for trialkoxysilane in particular, especially carbon numbers, such as methoxysilane, have an alkoxyl group (methoxy group) of 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99, 100, 101, 102, 103, 104, 105, 106, 107, 108, 109, 110, 111, 112, 113, 114, 115, 116, 117, 118, 119, 120, 121, 122, 123, 124, 125, 126, 127, 128, 129, 130, 131, 132, 133, 134, 135, 136, 137, 138, 139, 140, 141, 142, 143, 144, 145, 146, 147, 148, 149, 150, 151, 152, 153, 154, 155, 156, 157, 158, 159, 160, 161, 162, 163, 164, 165, 166, 167, 168, 169, 170, 171, 172, 173, 174, 175, 176, 177, 178, 179, 180, 181, 182, 183, 184, 185, 186, 187, 188, 189, 190, 191, 192, 193, 194, 195, 196, 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797, 798, 799, 800, 801, 802, 803, 804, 805, 806, 807, 808, 809, 810, 811, 812, 813, 814, 815, 816, 817, 818, 819, 820, 821, 822, 823, 824, 825, 826, 827, 828, 829, 830, 831, 832, 833, 834, 835, 836, 837, 838, 839, 840, 841, 842, 843, 844, 845, 846, 847, 848, 849, 850, 851, 852, 853, 854, 855, 856, 857, 858, 859, 860, 861, 862, 863, 864, 865, 866, 867, 868, 869, 870, 871, 872, 873, 874, 875, 876, 877, 878, 879, 880, 881, 882, 883, 884, 885, 886, 887, 888, 889, 890, 891, 892, 893, 894, 895, 896, 897, 898, 899, 900, 901, 902, 903, 904, 905, 906, 907, 908, 909, 910, 911, 912, 913, 914, 915, 916, 917, 918, 919, 920, 921, 922, 923, 924, 925, 926, 927, 928, 929, 930, 931, 932, 933, 934, 935, 936, 937, 938, 939, 940, 941, 942, 943, 944, 945, 946, 947, 948, 949, 950, 951, 952, 953, 954, 955, 956, 957, 958, 959, 960, 961, 962, 963, 964, 965, 966, 967, 968, 969, 970, 971, 972, 973, 974, 975, 976, 977, 978, 979, 980, 981, 982, 983, 984, 985, 986, 987, 988, 989, 990, 991, 992, 993, 994, 995, 996, 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1164, 1165, 1166, 1167, 1168, 1169, 1170, 1171, 1172, 1173, 1174, 1175, 1176, 1177, 1178, 1179, 1180, 1181, 1182, 1183, 1184, 1185, 1186, 1187, 1188, 1189, 1190, 1191, 1192, 1193, 1194, 1195, 1196, 1197, 1198, 1199, 1200, 1201, 1202, 1203, 1204, 1205, 1206, 1207, 1208, 1209, 1210, 1211, 1212, 1213, 1214, 1215, 1216, 1217, 1218, 1219, 1220, 1221, 1222, 1223, 1224, 1225, 1226, 1227, 1228, 1229, 1230, 1231, 1232, 1233, 1234, 1235, 1236, 1237, 1238, 1239, 1240, 1241, 1242, 1243, 1244, 1245, 1246, 1247, 1248, 1249, 1250, 1251, 1252, 1253, 1254, 1255, 1256, 1257, 1258, 1259, 1260, 1261, 1262, 1263, 1264, 1265, 1266, 1267, 1268, 1269, 1270, 1271, 1272, 1273, 1274, 1275, 1276, 1277, 1278, 1279, 1280, 1281, 1282, 1283, 1284, 1285, 1286, 1287, 1288, 1289, 1290, 1291, 1292, 1293, 1294, 1295, 1296, 1297, 1298, 1299, 1300, 1301, 1302, 1303, 1304, 1305, 1306, 1307, 1308, 1309, 1310, 1311, 1312, 1313, 1314, 1315, 1316, 1317, 1318, 1319, 1320, 1321, 1322, 1323, 1324, 1325, 1326, 1327, 1328, 1329, 1330, 1331, 1332, 1333, 1334, 1335, 1336, 1337, 1338, 1339, 1340, 1341, 1342, 1343, 1344, 1345, 1346, 1347, 1348, 1349, 1350, 1351, 1352, 1353, 1354, 1355, 1356, 1357, 1358, 1359, 1360, 1361, 1362, 1363, 1364, 1365, 1366, 1367, 1368, 1369, 1370, 1371, 1372, 1373, 1374, 1375, 1376, 1377, 1378, 1379, 1380, 1381, 1382, 1383, 1384, 1385, 1386, 1387, 1388, 1389, 1390, 1391, 1392, 1393, 1394, 1395, 1396, 1397, 1398, 1399, 1400, 1401, 1402, 1403, 1404, 1405, 1406, 1407, 1408, 1409, 1410, 1411, 1412, 1413, 1414, 1415, 1416, 1417, 1418, 1419, 1420, 1421, 1422, 1423, 1424, 1425, 1426, 1427, 1428, 1429, 1430, 1431, 1432, 1433, 1434, 1435, 1436, 1437, 1438, 1439, 1440, 1441, 1442, 1443, 1444, 1445, 1446, 1447, 1448, 1449, 1450, 1451, 1452, 1453, 1454, 1455, 1456, 1457, 1458, 1459, 1460, 1461, 1462, 1463, 1464, 1465, 1466, 1467, 1468, 1469, 1470, 1471, 1472, 1473, 1474, 1475, 1476, 1477, 1478, 1479, 1480, 1481, 1482, 1483, 1484, 1485, 1486, 1487, 1488, 1489, 1490, 1491, 1492, 1493, 1494, 1495, 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1662, 1663, 1664, 1665, 1666, 1667, 1668, 1669, 1670, 1671, 1672, 1673, 1674, 1675, 1676, 1677, 1678, 1679, 1680, 1681, 1682, 1683, 1684, 1685, 1686, 1687, 1688, 1689, 1690, 1691, 1692, 1693, 1694, 1695, 1696, 1697, 1698, 1699, 1700, 1701, 1702, 1703, 1704, 1705, 1706, 1707, 1708, 1709, 1710, 1711, 1712, 1713, 1714, 1715, 1716, 1717, 1718, 1719, 1720, 1721, 1722, 1723, 1724, 1725, 1726, 1727, 1728, 1729, 1730, 1731, 1732, 1733, 1734, 1735, 1736, 1737, 1738, 1739, 1740, 1741, 1742, 1743, 1744, 1745, 1746, 1747, 1748, 1749, 1750, 1751, 1752, 1753, 1754, 1755, 1756, 1757, 1758, 1759, 1760, 1761, 1762, 1763, 1764, 1765, 1766, 1767, 1768, 1769, 1770, 1771, 1772, 1773, 1774, 1775, 1776, 1777, 1778, 1779, 1780, 1781, 1782, 1783, 1784, 1785, 1786, 1787, 1788, 1789, 1790, 1791, 1792, 1793, 1794, 1795, 1796, 1797, 1798, 1799, 1800, 1801, 1802, 1803, 1804, 1805, 1806, 1807, 1808, 1809, 1810, 1811, 1812, 1813, 1814, 1815, 1816, 1817, 1818, 1819, 1820, 1821, 1822, 1823, 1824, 1825, 1826, 1827, 1828, 1829, 1830, 1831, 1832, 1833, 1834, 1835, 1836, 1837, 1838, 1839, 1840, 1841, 1842, 1843, 1844, 1845, 1846, 1847, 1848, 1849, 1850, 1851, 1852, 1853, 1854, 1855, 1856, 1857, 1858, 1859, 1860, 1861, 1862, 1863, 1864, 1865, 1866, 1867, 1868, 1869, 1870, 1871, 1872, 1873, 1874, 1875, 1876, 1877, 1878, 1879, 1880, 1881, 1882, 1883, 1884, 1885, 1886, 1887, 1888, 1889, 1890, 1891, 1892, 1893, 1894, 1895, 1896, 1897, 1898, 1899, 1900, 1901, 1902, 1903, 1904, 1905, 1906, 1907, 1908, 1909, 1910, 1911, 1912, 1913, 1914, 1915, 1916, 1917, 1918, 1919, 1920, 1921, 1922, 1923, 1924, 1925, 1926, 1927, 1928, 1929, 1930, 1931, 1932, 1933, 1934, 1935, 1936, 1937, 1938, 1939, 1940, 1941, 1942, 1943, 1944, 1945, 1946, 1947, 1948, 1949, 1950, 1951, 1952, 1953, 1954, 1955, 1956, 1957, 1958, 1959, 1960, 1961, 1962, 1963, 1964, 1965, 1966, 1967, 1968, 1969, 1970, 1971, 1972, 1973, 1974, 1975, 1976, 1977, 1978, 1979, 1980, 1981, 1982, 1983, 1984, 1985, 1986, 1987, 1988, 1989, 1990, 1991, 1992, 1993, 1994, 1995, 1996, 1997, 1998, 1999, 2000, 2001, 2002, 2003, 2004, 2005, 2006, 2007, 2008, 2009, 2010, 2011, 2012, 2013, 2014, 2015, 2016, 2017, 2018, 2019, 2020, 2021, 2022, 2023, 2024, 2025, 2026, 2027, 2028, 2029, 2030, 2031, 2032, 2033, 2034, 2035, 2036, 2037, 2038, 2039, 2040, 2041, 2042, 2043, 2044, 2045, 2046, 2047, 2048, 2049, 2050, 2051, 2052, 2053, 2054, 2055, 2056, 2057, 2058, 2059, 2060, 2061, 2062, 2063, 2064, 2065, 2066, 2067, 2068, 2069, 2070, 2071, 2072, 2073, 2074, 2075, 2076, 2077, 2078, 2079, 2080, 2081, 2082, 2083, 2084, 2085, 2086, 2087, 2088, 2089, 2090, 2091, 2092, 2093, 2094, 2095, 2096, 2097, 2098, 2099, 2100, 2101, 2102, 2103, 2104, 2105, 2106, 2107, 2108, 2109, 2110, 2111, 2112, 2113, 2114, 2115, 2116, 2117, 2118, 2119, 2120, 2121, 2122, 2123, 2124, 2125, 2126, 2127, 2128, 2129, 2130, 2131, 2132, 2133, 2134, 2135, 2136, 2137, 2138, 2139, 2140, 2141, 2142, 2143, 2144, 2145, 2146, 2147, 2148, 2149, 2150, 2151, 2152, 2153, 2154, 2155, 2156, 2157, 2158, 2159, 2160, 2161, 2162, 2163, 2164, 2165, 2166, 2167, 2168, 2169, 2170, 2171, 2172, 2173, 2174

[0142]

[Formula 2]



[0143]

as is mentioned. The principal chain skeleton of a polyoxyalkylene series polymer may consist of one kind of repeating unit, and may consist of two or more kinds of repeating units. When used especially for sealant etc., it is desirable from that what comprises the polymer which use a propylene oxide polymer as the main ingredients is amorphous, or the point which is hypoviscosity comparatively.

[0144]

As a synthetic method of a polyoxyalkylene series polymer. For example, the polymerizing method by an alkali catalyst like KOH, the polymerizing method by a transition metal compound and porphyrin complex catalyst like a complex produced by making an organoaluminum compound and porphyrin which are shown in JP-61-215623.A, JP-46-27508.B, JP-46-27509.B, JP-49-15330.B, a U.S. Pat. No. 3278457 item, a U.S. Pat. No. 3278458 item, a U.S. Pat. No. 3278459 item, a U.S. Pat. No. 3427256 item, a U.S. Pat. No. 3427334 item, the polymerizing method by a composite metal cyanide complex catalyst shown in a U.S. Pat. No. 3427335 item etc., \*\*\*\*\*\*, such as the polymerizing method using a catalyst which consists of a polyphosphazene salt illustrated by JP-H10-273512.A, and the polymerizing method using a catalyst which consists of a phosphazene compound illustrated by JP-H11-060722.A, are not limited in particular.

[0145] The polymerizing method of a polyoxyalkylene series polymer which has a reactive silicon group. JP-45-26319.B, JP-50-12154, JP-50-15693.A, JP-46-0906, JP-13707, 55-13468. What is proposed by each patent, such as JP-164123, JP-23-2450.B, U.S. Pat. No. 3632557, U.S. Pat. No. 4245063, U.S. Pat. No. 4369307, and U.S. Pat. No. 4960844, JP-61-17631.A, 61-215622, 61-215623. Although 6,000 or more number average molecular weights and Mw/Mn which are proposed by each patent of 61-218632, JP-75-72527.A, JP-73-47825.A, and JP-78-231707.A can illustrate a polyoxyalkylene series polymer with narrow molecular weight distribution in the 1.6 or less amount of Polymer Division, it is not limited to in particular these.

[0146]

A polyoxyalkylene series polymer which has the above-mentioned reactive silicon group may be used alone, and may be used together two or more sorts.

[0147]

Said saturated hydrocarbon system polymer is a polymer which does not contain substantially carbon-carbon unsaturated bonds other than of the aromatic ring, and a polymer which makes the skeleton, (1) whether an olefinic compound of the carbon numbers 1-6, such as ethylene, propylene, 1-butene, and isobutylene, is polymerized as a main monomer, and (2) After making diene series, such as butadiene and isoprene, homopolymerize or carry out copolymerization of the above-mentioned olefinic compound, can obtain by a method of hydrogenating, but. Since an isobutylene system polymer and a hydrogenation polybutadiene system polymer tend to introduce a functional group into an end, and tend to control a molecular weight and can increase the number of functional groups, they are preferred, and a composite case to especially their isobutylene system polymer is preferred.

[0148]

That whose principal chain skeleton is a saturated hydrocarbon system polymer has the feature which is excellent in heat resistance, weatherability, endurance, and humidity interception nature.

[0149]

All the monomeric units may be formed from an isobutylene unit, and an isobutylene system polymer. Although a copolymer with other monomers may be sufficient, what contains a repeating unit which is generated from a fluid of rubber property 50% of the weight or more is preferred, what is generated from a fluid of rubber property 80% of the weight or more is preferred, and especially a thing contained 90 to 95% of the weight is preferred.

[0150]

As a synthetic method of a saturated hydrocarbon system polymer, although various polymerization methods are reported conventionally, especially living polymerization what is called of recent years many is developed, an higher polymerization (I. — P.Kennedy et al.) which was found out by Kennedy and others in the case of a saturated hydrocarbon system polymer, especially an isobutylene system polymer J. Polymer Sci. and Polymer Chem. Ed. 1997, By using 15 volumes and 2843 pages, manufacturing easily is possible, about 500 to 100,000 molecular weight can be polymerized in 1.5 or less molecular weight distribution, and it is known that various functional groups can be introduced into a molecular terminal.

[0151]

As a process of a saturated hydrocarbon system polymer which has a reactive silicon group. For example, JP-4-68693.B, JP-7-108228.B, JP-63-254149.A, Although it writes in each Description of JP-64-22904.A, JP-1-01509.A, Patent Gazette No. 2539445, Patent Gazette No. 2873395, and JP-7-33852.A, it is not limited to in particular these.

[0152]

It is a general formula in a saturated hydrocarbon system polymer which has the above-mentioned reactive silicon group (6):



Especially (A7) a saturated hydrocarbon system polymer that has a basis expressed with (R<sub>4</sub>in formula is the same as the above) can be used as an ingredient. This (A7) ingredient has the feature which is excellent in heat resistance based on a saturated hydrocarbon system polymer, weatherability, and humidity interception nature of a principal chain skeleton, and does not have generation of methanol accompanying a hydrolysis reaction of a reactive silicon group, and is a polymer whose stability of a hardened material, endurance, and creep resistance are still better.

[0153]

A saturated hydrocarbon system polymer which has the above-mentioned reactive silicon group may be used alone, and may be used together two or more sorts.

[0154]

Especially in this invention, a chain can use what is an acryloxy ester (meta) system copolymer as an ingredient (A8) in an organic polymer of the (A) ingredient.

[0155]

Especially as an acryloxy ester (meta) system monomer which constitutes a main chain of the above-mentioned acryloxy ester system polymer, it is not limited but various kinds of things can be used. If it illustrates, acryloxy acid (meta), methyl acrylate (meta), (Meta) Ethyl acrylate, acrylate acid (meta)-propyl, acryloxy acid (meta) isopropyl, (Meta) Acrylic acid-n-butyl, isobutyl acrylate (meta), (Meta) Acrylic acid-tert-butyl, acryloxy acid (meta)-pentyl, (Meta) Acrylic acid-n-hexyl, acryloxy acid-2-methyl cyclohexyl, (Meta) Acrylic acid-n-heptyl, acryloxy acid (meta)-n-octyl, (Meta) Acrylic acid-2-ethylhexyl, acryloxy acid (meta) nonyl, (Meta) Decyl acrylate, acryloxy acid (meta) dodecyl, acryloxy acid phenyl, (Meta) Acrylic acid tolyl, acryloxy acid (meta) benzyl, acryloxy acid (meta)-2-methoxy ethyl, (Meta) Acrylic acid-3-methoxy butyl, acryloxy acid (meta)-2-hydroxyethyl, (Meta) Acrylic acid-2-hydroxypropyl, acryloxy acid (meta) steryl, methylglycidyl acrylate (meta), acryloxy acid (meta)-2-aminoethyl, gamma-(methacryloyl oxypropyl) trimethoxysilane, an ethylenedioxy addition of acryloxy acid (meta), (Meta) Acrylic acid trifluoromethyl thioether, an acryloxy acid (meta)-2-trifluoro methylthyl, (Meta) Acrylic acid 2-perfluoro ethyl, acryloxy acid (meta)-2-perfluoro ethyl-2-perfluoro butylthyl, (Meta) Acrylic acid 2-perfluoro ethyl, acryloxy acid (meta) perfluoro methyl, (Meta) Acrylic acid JPA fluoromethylthyl, acryloxy acid (meta)-2-perfluoro methyl-2-perfluoro ethylthyl, (Meta) Acrylic acid JPA fluoromethylthyl, acryloxy acid (meta)-2-perfluoro hexylethyl, acryloxy acid (meta)



group, a saturated hydrocarbon system polymer which has a reactive silicon group, an acryloyl ester system copolymer which has a reactive silicon group (meta), and an organic polymer which blends two or more sorts chosen from a group, \* and others, can also be used.

[0165]

A manufacturing method of an organic polymer, which blends a polyoxallylene series polymer which has a reactive silicon group, and an acryloyl ester system copolymer which has a reactive silicon group (meta), Although proposed by JP-59-122541A, JP-59-12642A, JP-H6-172831A, JP-H11-116165A, etc., it is not limited to in particular these.

[0166]

It is known as compared with a case where a polyoxallylene series polymer is independently used alone, that the polymer which blends a polyoxallylene series polymer which has its reactive silicon group, and an acryloyl ester system copolymer which has a reactive silicon group (meta) that stability is bid. Then the above-mentioned general formula (6) as a polyoxallylene series polymer component in the aforementioned organic polymer to blend:

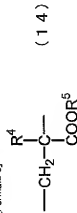


A polyoxallylene series polymer (A5) which has a basis expressed with  $(\text{R}^1)_n$  formula is the same as the above) is used. An organic polymer blended with an acryloyl ester system copolymer (A6) which has a reactive silicon group (meta) has outstanding stability, endurance, and creep resistance based on the (A5) ingredient, while outstanding weatherability and an adhesive property based on an ingredient (A6) are shown.

[0167]

(A6) A desirable example of an acryloyl ester (meta) system copolymer of an ingredient has a reactive silicon group, and a chain is a following general formula substantially (14):

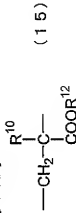
[Formula 3]



[0169]

The acryloyl ester monomer unit which has an alkyl group of the carbon numbers 1-8 expressed with (a hydrogen atom or a methyl group, and  $\text{R}^1$  show the alkyl group of the carbon numbers 1-8 among a formula, as for  $\text{R}^{10}$  (meta), and following general formula (15):

[Formula 4]



[0171]

(— inside of formula, and  $\text{R}^{10}$  — the above — the same —  $\text{R}^{12}$  shows a with a carbon numbers of ten or more carbon group —) to a copolymer which consists of an acryloyl ester monomer unit which has a with a carbon numbers of ten or more expressed alkyl group (meta-7). It is the method of blending and manufacturing a polyoxallylene series polymer which has a reactive silicon group. [0172]

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as  $\text{R}^1$  of said general formula (14) — the carbon numbers 1-8 of a methyl group, an ethyl group, a propyl group, n-butyl group, t-butyl group, a 2-ethylhexyl group, etc. — desirable — 1-4 — an alkyl group of 1-2 is raised still more preferably. An alkyl group of  $\text{R}^1$  may be independent and may be mixed two or more sorts.

[0173]

as  $\text{R}^2$  of said general formula (15) — ten or more carbon numbers of a lauryl group, a tridecyl group, a cetyl group, a stearyl group, a behenyl group, etc. — usually — 10-30 — a long-chain alkyl group of 10-20 is raised preferably. Like a case of  $\text{R}^1$ , an alkyl group of  $\text{R}^2$  may be independent and may be mixed two or more sorts.

[0174]

Although a chain of a \* (meta) acryloyl ester system copolymer consists of a monomeric unit of a formula (14) and a formula (15) substantially, a "real target" here means that the sum total of a monomeric unit of a formula (14) which exists in this copolymer, and a formula (15) supposes 80 % of the weight. The sum total of a monomeric unit of a formula (14) and a formula (15) is 70 % of the weight or more preferably.

[0175]

As for an abundance ratio of a monomeric unit of a formula (14), and a monomeric unit of a formula (15), 95:5-40:60 are preferred at a weight ratio, and 90:10-60:40 are still more preferred.

[0176]

As monomeric units other than a formula (14) which may be contained in this copolymer, and a formula (15). For example, acrylic acid, such as acryloyl acid and methacrylic acid; Acrylamide, Amide groups, such as methacrylamide, N-methylacrylamide, and N-methylmethacrylamide; Epoxy groups, such as glycidyl acrylate and glycidyl methacrylate; Diethylamino ethyl acrylate, diethylamino ethyl methacrylate. A monomer containing amino group, such as acryloylamine, methacryloylamine, and methacryloylamine; and other monomers, such as allyl vinyl ether, VCM(PVC, vinyl acetate), vinyl propionate, ethylene, etc. in addition to this is raised.

[0177]

Although an organic polymer which blends a saturated hydrocarbon system polymer which has a reactive silicon group, and an acryloyl ester system copolymer which has a reactive silicon group (meta) is proposed by JP-H1-18876A, JP-2000-186176A, etc., it is not limited to in particular these.

[0178]

A method of polymerizing an acryloyl ester (meta) system monomer elsewhere under existence of an organic polymer which has a reactive silicon group as a manufacturing method of an organic polymer which blends an acryloyl ester system copolymer which has a reactive silicon functional group (meta) can be used. Although this manufacturing method is concretely indicated by each gazette, such as JP-59-78223A, JP-59-168014A, JP-60-228516A, JP-60-228517A, it is not limited to these.

[0179]

In the invention, silica can be used as a (B) ingredient. This silica has the function to improve the stability of an organic polymer which is the (A) ingredient of this invention, endurance, and creep resistance.

[0180]

(C) Silicate which is an ingredient is a general formula (16).



( $\text{R}^{13}$  are a hydrogen atom or an alkyl group of the carbon numbers 1-20, an aryl group of the carbon numbers 6-20, and the univalent hydrocarbon group chosen from an aralkyl group of the carbon numbers 7-20 independently among a formula, respectively) — they are tetraalkoxysilane expressed or to partial hydrolysis condensate.

[0181]

As an example of silicate, for example A tetramethoxy silane, a tetraethoxysilane, Ethoxy trimethoxysilane, dimethoxy diethoxysilane, methoxy triethoxysilane, Tetraalkoxysilane (tetraalkyl silicates), such as tetra n-propoxysilane, tetra n-butoxysilane, tetra t-butoxysilane, tetra t-butoxysilane, and tetra t-butoxysilane, and those partial hydrolysis condensates are raised. [0182]

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[0183]

[0184]

[0185]

alliance may be used only by the kind, and may carry out one or more of their own affairs.

[0186]

[0187]

[8810]

[0189]

acid. Straight chain saturated fatty acid groups, such as RAKUSERU acid; Undecylenic acid, Linder

[illegible]

16, 19-docosatetraenoic acid, 4,8,12,15,18-eicosapentaenoic acid, clupanodonic acid, clupanodonic acid, herring acid,

malvallic acid, sterculic acid, HIDONO carbonyl acid, Alicyclic carboxylic acids, such

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cerebronic acid; a halogenation product of monocarboxylic acid, such as chloracetic acid, 2-

acid, citrate, and isocitric acid, etc. are mentioned. As aromatic carboxylic acid, benzoic acid, 9-

arginine, cysteine, methionine, phenylalanine, tryptophan, and histidine, is mentioned.

when the melting point of acid carboxylic acid is high (crystallinity is high), the melting point becomes

[0192]

state of viscosity and which is hard to deal with it (workability — bad). On the contrary, when a certain number of acid carboxylic acid is small (a molecular weight is small) catalysability of

acid metal salt may fall greatly. Therefore, as for said carboxylic acid, it is preferred that carbon

carboxylic acid tin salt to dicarboxylic acid, and it is more preferred that it is

(ii)  $\sum_{n=0}^{\infty} \frac{1}{n!} = e$

(It is the same as the above the inside R of a formula.) Two RCOO-bases may be the same and may

carboxylic acid will salt (free decanoic acid and in which said carboxylic acid anion (C) is an anion of a carboxylic acid (2-methylsuccinic acid, etc.) and the 4th class cation whose cation of an alpha

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[0196]

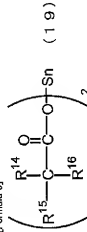
Especially in this invention, carbon of an alpha position of a carboxyl group used as a carboxylic acid tin salt which is the 4th class carbon as an ingredient (C1) in carboxylic acid tin salt (Q).

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(C1) A carboxylic acid tin salt of an inorganic acid is a general formula (19).

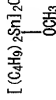
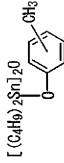
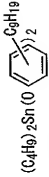
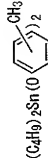
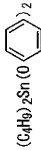
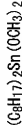
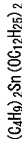
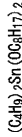
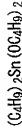
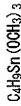
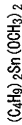
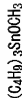
[0199g]

[Formula 5]









[0225]

Although \*\* is mentioned, it is not limited to these. In these, a dialkyl tin JIARUKOKI acid is preferred. Especially the dibutyl tin JIMETOKI acid is low cost, and since it is easy to receive, it is preferred.

[0226]

(D) As amount of ingredient used, about 0.01–20 weight sections are preferred to ingredient (A1) 100 weight section. Also about 0.1–10 weight sections are preferred to ingredient (E) 100 weight section. Since the amount of ingredient used will become difficult to advance, if loadings are less than this range, it is not desirable. On the other hand, if loadings exceed this range, working life becomes short too much, and workability may worsen, and it is not desirable from a point of storage stability.

[0227]

As amount of [ in case used of using the (D) ingredient and the (C) ingredient together as a curing catalyst 1, (A1) it is preferred to consider it as (C) ingredient 0.5 – 20 weight section and (D) ingredient 0.01 – 10 weight section to ingredient 100 weight section, and also it is more preferred to consider it as (C) ingredient 1 – 10 weight section and (D) ingredient 0.02 – 5 weight section. (C) When loadings of an ingredient are less than this range, a cure rate may become slow, when loadings exceed this range, working life becomes short too much and workability may worsen. (D) The stability of a hardened material which will be obtained if an improvement effect of hardenability, depth of hardenability, an adhesive property, and thin layer hardenability may not be enough if loadings of an ingredient are less than this range, and loadings exceed this range, endurance, and creep resistance may worsen.

[0228]

The (D) ingredient can be used combining two or more sorts besides using it alone.

[0229]

In this invention, a non-tin catalyst can be used as a (E) ingredient. This non-tin catalyst has a function which improves the stability of hardened material obtained, endurance, and creep resistance. It is combined with other cured hardening catalysts, when it uses as a radical condensation catalyst of an organic polymer which is an ingredient (A1) of this invention. A non-tin catalyst which is the (E) ingredient is an eco-friendly curing catalyst with high social needs.

[0230]

As a non-tin catalyst which is the (E) ingredient which can be used for this invention, although there is no restriction in particular, an organic metallic compound containing carboxylic acid, carboxylic acid metal salt other than carboxylic acid tin salt, organic sulfonic acid, alkyl acid phosphate and 3B fellows, and 4A group metal, etc. are illustrated.

[0231]

The various above-mentioned carboxylic acid which has an acid radical of carboxylic acid tin salt which is the (C) ingredient as carboxylic acid can be illustrated.

[0232]

As for said carboxylic acid, it is preferred like carboxylic acid tin salt (C) that carbon numbers including carbon of a carbonyl group are 2–20. It is more preferred that it is 6–17, and it is preferred that it is especially 8–12. A point to dicarboxylic acid or monocarboxylic acid of this ester (workability, viscosity) of dealing with it of carboxylic acid is preferred, and monocarboxylic acid is more preferred. Carboxylic acid (n-octadecanoic acid). In which said carboxylic acid is carboxylic acid (2-ethyl-2-methyl-2-octadecanoic acid etc.) and the 4th class carbon whose carbon of an alpha position of a carboxylic acid is the 4th class carbon A phthalic acid etc. are more preferred from a cure rate, but quickness of workability carboxylic acid whose carbon atom which adjoins a carbonyl group is the 4th class carbon is preferred.

[0233]

Especially as carboxylic acid, 2-ethylhexanoic acid, n-octadecanoic acid, BASA tic acid, 2,2-dimethyloctanoic acid and 2-ethyl-2,5-dimethylhexanoic acid are preferred from a point of availability, hardenability, and workability.

[0234]

As carboxylic acid metal salt other than said carboxylic acid tin salt, metal salt of the various above-mentioned carboxylic acid can be used conveniently.

[0235]



[0247]

By carboxylic acid metal salt other than carboxylic acid tin salt, organic sulfonic acid, and alkyl acid phosphate, activity is low, and when moderate hardenability is not acquired, an amine compound can be added as a co-catalyst.

[0248]

As various amine compounds, the indicated various above-mentioned amine compounds can be used as a co-catalyst of carboxylic acid tin salt (C).

[0249]

Loadings of said amine compound, about 0.01–20 weight sections are preferred to organic polymer (100 weight section of an ingredient (A1)), and also its 0.1 to 5 weight section is more preferred. A cure rate may become a little that loadings of an amine compound are less than 0.01 weight sections, and a hardening reaction becomes fully difficult to advance. On the other hand, if loadings of an amine compound exceed 20 weight sections, pot life may become short too much and is not preferred from a point of workability.

[0250]

As a metal system compound of non-tin, besides carboxylic acid metal salt other than said carboxylic acid tin salt, An organic metallic compound containing 3B fellows and 4A group metal is raised, and although a titanate organic compound, an organoaluminum compound, an organic zirconium compound, an organic boron compound, etc. are preferred from a point of activity, it is not limited to these.

[0251]

As said titanate organic compound, tetraisopropyl titanate, *tert*-butyl titanate, tetramethyl titanate, *tert*-butyl titanate, titanate compound, such as titanium chelate, such as titanium alkoxides, such as triethanolamine titanate, titanium triacetylacetonate, titanium ethylacetylacetonate, ethylene glycolate, and titanium lactate, etc. are raised.

[0252]

As said organoaluminum compound, aluminum isopropylate, aluminum alkoxides, such as mono-*sec*-butoxy aluminum diisopropylate and aluminum *sec*-butoxylate. Aluminum chelate, such as aluminum triacetylacetonate, aluminum triacetylacetonate, and diisopropyl aluminum ethylacetylacetonate is

[0253]

As said zirconium compound, zirconium tetra isopropoxide, zirconium alkoxides, such as a zirconium tetra-*n*-butoxy, zirconium normal butyrate, zirconium chelate, such as zirconium tetra acetylacetonate, zirconium monoacetyl acetate, zirconium bisacetylacetonate, zirconium acetylacetonate bis-ethylacetylacetonate, and zirconium acetate, is raised.

[0254]

Although \*\*\* concomitant use is also possible so, these titanate organic compounds, an organoaluminum compound, an organic zirconium compound, an organic boron compound, etc., it is desirable in a viewpoint which can reduce the amount of catalyst used especially according to concomitant use with said amine compound or in alkyl-acid-phosphate compound since it is possible to improve activity, and more desirable in a viewpoint of adjustment of working life in hardenability and ordinary temperature in an elevated temperature.

[0255]

(E) As amount of ingredient used, about 0.01–20 weight sections are preferred to ingredient (A1) 100 weight section, and also about 0.5–10 weight sections are preferred. Since a cure rate may become slow and a hardening reaction will become fully difficult to advance if loadings are less than this range, it is not desirable. On the other hand, if loadings exceed this range, working life becomes short too much, and workability may worsen, and it is not desirable from a point of storage stability.

[0256]

The (E) ingredient can be used combining two or more sorts besides using it alone.

[0257]

In this invention, a minute hollow body can be used as a (F) ingredient. While improving the workability (\*\*\*\*\*, thiophony) of a constituent, notably as indicated to JP H11-35922A or JP H11-31072A (if this minute hollow body is used, it is known that a weight saving of a constituent and low-costing are possible. However, it is known that the stability of a hardened material of a hardenability

constituent and endurance which are obtained will fall according to an addition of this minute hollow body.

[0258]

This hardenability constituent, which added a minute hollow body of the (F) ingredient by using an organic polymer which is an ingredient (A1) of this invention as a polymer component can maintain highly the stability of a hardened material and endurance which are obtained, improving workability (\*\*\*\*\*) notably.

[0259]

A very small hollow body (one-fourth a balloon) which is the (F) ingredient of this invention is a hollow body by which a diameter was preferably constituted from material of minerals of 500 micrometers or less or quality of organic (F) 1 mm or less as indicated, for example on "state-of-the-art of a functional filler" (CMC). Of an ingredient in particular is not limited but it is [Ingredient.] usable in various kinds of publicly known balloons.

[0260]

As for average particle density of a balloon, it is preferred that it is 0.01–1.0g/cm<sup>3</sup>; it is more preferred that it is 0.05–0.7g/cm<sup>3</sup> and it is preferred that it is especially 0.1–0.5g/cm<sup>3</sup>. If tonnage strength of a hardened material may fall if average particle density is less than this range, and average particle density exceeds this range on the other hand, a workability improvement effect may not be enough.

[0261]

An inorganic system balloon is more preferred than a point of stability and endurance to an organic system balloon.

[0262]

As said inorganic system balloon, can illustrate a silicic acid system balloon and a non-silicic acid system balloon, fly ash balloons, etc. can illustrate an alumina balloon, a zirconia balloon, a carbon balloon, etc. on a non-silicic acid system balloons. As an example of these inorganic system balloons, as a mill balloon, a win light by JICHI Chemicals, As a SANDI light by Sand Engineering Co., Ltd. and glass balloons, KARIN by Nippon Sheet Glass Co., Ltd. The Sumitomo 3M cell star Z-28, MICRO BALLOON made from EMERSONCUMING, CELAMOD GLASSMODULES made from PITTSBURGE CORNING, AS GLASS BUBBLES made from 3M, FLUIDA tube made from Fuji SHIRISHA Chemicals, and a silica balloon, as G-CEL by Asahi Glass Co., Ltd. SAURASHA made from Fuji SHIRISHA Chemicals, and fly ash balloons, CEROSOL by BASF AG, and METEORING PLUTON by BASF AG. ZIRCONIUM SPIRALS made from BASF AG, BW by Shikoku K.K. and K.K. and K.K. and HOLLOW ZIRCONIUM SPIRALS made from ZIRCOA, KUREKASU fair made from Kurachi Chemicals and product are lots fair made from GENERAL TECHNOLOGIES are marketed as a carbon balloon.

[0263]

A balloon of thermosetting resin and a balloon of thermoplastics can be illustrated as said organic system balloon. On a thermosetting balloon, a phenol balloon, an epoxy balloon, and a urea balloon can illustrate a sarin balloon, a polystyrene balloon, a polymethacrylate balloon, a polyvinyl alcohol balloon, and a styrene acrylic balloon at a thermoplastic balloon. A balloon of thermoplastics which constructed the bridge can also be used. A balloon after foaming may be sufficient, and a balloon here is made to foam, after blending a thin containing a foaming agent, and is good also as a balloon.

[0264]

As an example of these organic system balloons, as a phenol balloon, Union Carbide UCAR and PHENOLIC MICROBALLOONS. As a balloon, EPOCOSPHERES made from EMERSONCUMING. As a urea balloon, EPOCOSPHERES VF-O made from EMERSONCUMING. As a sarin balloon, SARAN MICROSPHERES made from DOW CHEMICAL. Expanded made from Japanese filament, the Matsumoto Yushi-Seiyaku, Matsumoto microsphere. As a polystyrene balloon, DYLITE EXPANDABLE POLYSTYRENE made from ARCO POLYMERS, SBX83 by Japan Synthetic Rubber Co., Ltd. (P) are marketed by EXPANDABLE POLYSTYRENE SBX83 made from BASF WYANDOTTE. And constructed type styrene acrylic acid balloon of a bridge.

[0265]

The above-mentioned balloon may be used alone, and two or more kinds may be mixed and it may be used. The surface of these balloons Fatty acid, fatty acid ester, rosin. What was processed in order















methylsilicate 51 which were used in working examples 3-4 are a condensate of a tetraethoxysilane and a tetramethoxy silane, respectively, and showed the especially outstanding effect.

**(Synthetic example 2)**

Use polyoxypropylene

Una polyoxypropylene glycol of the molecular weight 2,000 [about 1,000] as an initiator, and the hydroxy-terminated polyoxypropylene oxide of the number average molecular weight 14,500 [about 1,500] produced by polymerizing propylene oxide in the zinc bis-oxanocarbamate glyma complex compound catalyst is used. Allyl and polypropylene oxide was obtained in the same procedure as the synthetic example 1. To use, allyl and polypropylene oxide, in the same procedure as the synthetic example 1, it was made to react to trimethoxysilane and the polyoxyallyl series polymer (A-2) which has an average of 13 trimethoxysilane groups at the end was obtained.

[0324]

(Synthetic example 3)

To the allyl end polypropylene oxide obtained in the synthetic example 2, in the same procedure as the synthetic example 1, it was made to react to triethoxysilane and the polyoxyalkylene series polymer (A-3) which has an average of 15 triethoxy silyl groups at the end was obtained.

[0325]

(Synthetic example 4)

To the allyl end polypropylene oxide obtained in the synthetic example 2, in the same procedure as the synthetic example 1, it was made to react to methyl dimethoxysilane and the polyoxyalkylene series polymer (A-4) which has an average of 1.5 methyl dimethoxy silyl groups at the end was obtained.

[0326]

(The reference examples 5-11 and comparative examples 3-5)

Organic polymer (A-2-4) 100 weight section which has the reactive silicon group obtained in the synthetic examples 2-4 according to the combination formula shown in Table 2. Surface treatment with silane coupling agent (A-1) 100 weight section, silane coupling agent (A-2) 100 weight section, colloidal calcium carbonates (product made from Silexsilite industry, Hakusaka CO) 120 weight section, titanium oxide (Ushihara Sando) made from Silexsilite industry, HA-820 20 weight section, CDDI-2 weight section, the amount part of thixotropic grant agent (made in Kusumoto Chemicals) 1 DISUPAOR 65000 duples, and light stabilizer (the Sanfco make) SANORU LS7701 weight section and an ultraviolet ray absorber (in The Specialty Chemicals), TORUKARU 3271 weight section, antioxidant (product made from Duchi Shinko Chemical Industry, NIPPON UNIOX) 1 weight section. The amount part of dehydroxy vinyltrimethoxysilane (Nippon, Uniox make, A-117) duples, adhesion grant agent A-bulky (aminethoxy)-gamma-aminopropyl trimethoxysilane (the Nippon Uniox make) A-120. This is weight section, number of copies given [silicate (added in a oil coat, methylsilicate 93)] 1 in 10 (total), curing catalyst (Urethane acryl transformation additive) (the Urethane acryl make, STANN BLU) of the (C) (D) (E) (F) (G) (H) (I) (J) (K) (L) (M) (N) (O) (P) (Q) (R) (S) (T) (U) (V) (W) (X) (Y) (Z) (AA) (AB) (AC) (AD) (AE) (AF) (AG) (AH) (AI) (AJ) (AK) (AL) (AM) (AN) (AO) (AP) (AQ) (AR) (AS) (AT) (AU) (AV) (AW) (AX) (AY) (AZ) (BA) (BB) (BC) (BD) (BE) (BF) (BG) (BH) (BI) (BJ) (BK) (BL) (BM) (BN) (BO) (BP) (BQ) (BR) (BS) (BT) (BU) (BV) (BW) (BX) (BY) (BZ) (CA) (CB) (CC) (CD) (CE) (CF) (CG) (CH) (CI) (CJ) (CK) (CL) (CM) (CN) (CO) (CP) (CQ) (CR) (CS) (CT) (CU) (CV) (CW) (CX) (CY) (CZ) (DA) (DB) (DC) (DD) (DE) (DF) (DG) (DH) (DI) (DJ) (DK) (DL) (DM) (DN) (DO) (DP) (DQ) (DR) (DS) (DT) (DU) (DV) (DW) (DX) (DY) (DZ) (EA) (EB) (EC) (ED) (EE) (EF) (EG) (EH) (EI) (EJ) (EK) (EL) (EM) (EN) (EO) (EP) (EQ) (ER) (ES) (ET) (EU) (EV) (EW) (EX) (EY) (EZ) (FA) (FB) (FC) (FD) (FE) (FF) (FG) 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[0327]

It examined by having pulled by the same method as the above-mentioned using the class product of Table 2, and was extended at the time of intensity (MPa) and Ebfracture at the time of M50:50% baseline modulus (MPa) and Tbfacture, and (%) was measured. A result is shown in Table 2.

[0328]

The recovery was measured by the same method as the above-mentioned using the class product of Table 2. However, the stretched state was fixed at 23 \*\* 100% for 24 hours this time, and the recovery was measured from the state which opened this wide at 23 \*\* and the marked line restored 1 hour afterward. A result is shown in Table 2.

[0320]

(Creep measurement using a shear sample)

The displacement difference with the 140-hour back immediately after creating the \*\* sample which

is not carried an area of 20 mm x 25 mm and 1 mm in thickness using the class product of Table 2, imposing 0.1 MPa load for what recuperated [ 23  $\pm$ 3  $\pm$ 50  $\pm$ s ], itself on the 4th in 60  $\pm$ s oven, and imposing load was measured. [ par day ] The displacement difference made 0 x for the thing below 0.4 mm, and the displacement difference carried out a thing of 0.4 mm or more. A result is shown in table 2.

[0330]

[Table 2]







組成 (重量部)		反活性ケイ素基の構造		参考例		比較例	
有機重合体	(A4) 成分	A-10	トリエチルシリル基	100	100	9	10
		A-1	トリエチルシリル基	100	100	9	10
充填材		Winofil SPM		120	120	120	100
可塑剤		RFK-2		20	20	20	20
チクソ性付与剤		Cravallac super		50	50	50	50
光安定剤		#1-MLS-770		5	5	5	5
紫外線吸収剤		FXE-Y327		1	1	1	1
酸化防止剤		15545SP		1	1	1	1
脱水剤		A-171		2	2	2	2
接着性付与剤		A-1100	トリエチルシリル基	3	3	3	3
		A-1120	トリエチルシリル基	3	3	3	3
硬化触媒		材スクリュー-220		2	2	2	2
復元率		(%)		87	85	25	25
クリープ (せん断)		(min)		○	○	×	×
皮張時間		(min)		20	15	5	15
		(min)		20	15	5	15

[0955]

If the aminosiilane which has a triethoxy silyl group which is the (C) ingredient as an adhesive grant

agent is combined with the end of the (A4) ingredient using the polymer which has a triethoxy silyl group as an organic polymer as shown in the reference example 17 of Table 5. Exceeding in stability and creep resistance, change of the skinning time in storage order is small, and storage stability is good.

[0956]

The reference examples 18 and the comparative examples 11-12. One of the polymers (A-2) 100 weight parts which has the triethoxy silyl group obtained in the synthetic example 2 according to the combination formula shown in Table 6, as DDP20, weight section and a dehydrator — triethoxysilane (made in a oil coat) the N-beta-(aminomethyl)-gamma-aminopropyl trimethoxysilane (the Nippon Unicar make) which is the (H) ingredient as the amount part of ethyl silicate 28 duplex, and an adhesion grant agent A-1120 or N-beta-(aminomethyl)-gamma-aminopropyl triethoxysilane (the Shin-Etsu Chemical make). It added, KBE-4033 weight transformation make, neo SUTAN U-220 duplex were sealed in the glassware which carried out the nitrogen purge, and 1 liquid mold-curing nature constituent was obtained. In the comparative example 11, the leather-covered time test was performed under 50% of 23 ° humidity RH conditions, without the comparative example 12, after promoting the ester exchange reaction between reactive silyl groups by recuperating oneself for seven days at 50 ° in these 1 liquid mold-curing nature constituents, the leather-covered time test was performed under 50% of 23 ° humidity RH conditions. A result is shown in Table 6.

[0957]

[Table 6]



and a photo-setting resin (the Toagosei make), ARONIKUSU M-3053 weight section, light stabilizer (Sanjyo make, SANORULST701) 1 weight section, Ultraviolet ray absorbent (made in [The Specialty Chemicals] 1, thavin 327) 1 weight section, Zero copy of minute hollow body (the product made from the Fuji SHIRISHA chemicals, the FUJIBA line H-40 which is antioxi-dant (made in [The Specialty Chemicals] 1, IRUGA NOX 1010) 1 weight section and the (F) ingredient, or 20 copies were added, respectively, and it then kneaded with a 3 point roll, and was considered its base resin. After that added 20 copies of minute hollow bodies was made into the reference example 21, using (A-12) as an organic polymer. What added zero copy of minute hollow body was made into the comparative example 14, using (A-12) as an organic polymer, and what added 20 copies of minute hollow bodies was made into the comparative example 15, using (A-11) as an organic polymer.

[0364] Using the mixture of 2-ethylhexanoic acid tin (Japanese east transformation make, U-28) (divalent) 3 weight section, amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section as a hardening agent, above-mentioned base resin and hardening agent were mixed uniformly, and workability (\*\*\*\*\*), and endurance were evaluated.

[0365] The constituent of the reference example 21 had workability better than the comparative example 14, and its endurance was better than the comparative example 15.

[0366] (The reference example 22 and the comparative example 16)

Organic polymer (A-1) 95 weight section which has the reactive silicon group obtained in organic polymer (A-10) 70 weight section, which has the reactive silicon group obtained in the synthetic example 10, or the synthetic example 1, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, Hakukenka COR) 80 weight section, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, BUSUKO light R) 60 weight section, Heavy-calcium-carbonate (product made from Shiraishi industry, HOWATON SD) 20 weight section, DOP-40 weight section, epoxy system plasticizer (New Japan Chemical make, SANSON sizer EP-S) 20 weight section, thixotropic grant agent (made in [Kusumoto Chemicals] 1, DISUPARON 305) 3 weight section, and a photo-setting resin (the Toagosei make), ARONIKUSU M-3093 weight section, light stabilizer (Sanjyo make, SANORULST701) 1 weight section, Ultraviolet ray absorbent (made in [The Specialty Chemicals] 1, thavin 327) 1 weight section were measured, respectively, and it then kneaded with a 3 point roll, and was considered as base resin. What added 95 copies of (A-1) as an organic polymer was made into the reference example 22, and what added 95 copies of (A-1) as an organic polymer was made into the comparative example 16. As a hardening agent, to this base resin, the mixture of 2-ethylhexanoic acid tin (Japanese east transformation make, U-28) (divalent) 3 weight section, and amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section was added, and the recovery was measured to it.

[0367] The constituent of the reference example 22 showed the recovery higher than the comparative example 16, stopping weight % of an organic polymer low.

[0368] (The reference example 23 and the comparative example 17)

Organic polymer (A-10) 95 weight section which has the reactive silicon group obtained in the synthetic example 10, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, Hakukenka COR) 80 weight section, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, BUSUKO light R) 60 weight section, Heavy-calcium-carbonate (product made from Shiraishi industry, HOWATON SD) 20 weight section, DOP-40 weight section, thixotropic system plasticizer (New Japan Chemical make, SANSON sizer EP-S) 20 weight section, thixotropic grant agent (made in [Kusumoto Chemicals] 1, DISUPARON 305) 3 weight section, and a photo-setting resin (the Toagosei make), ARONIKUSU M-3093 weight section, light stabilizer (Sanjyo make, SANORULST701) 1 weight section, Ultraviolet ray absorbent (made in [The Specialty Chemicals] 1, thavin 327) 1 weight section, Antioxidant (made in [The Specialty Chemicals] 1, IRUGA NOX 1010) 1 weight section and zero copy of epoxy resin (the product made from Japan epoxy resin, Epicoat 828), or five copies were measured, respectively, and it then kneaded with a 3 point roll, and

was considered as base resin. What added five copies of epoxy resins was made into the reference example 23. What added zero copy of epoxy resin was made into the comparative example 17. As a hardening agent, to this base resin, the mixture of 2-ethylhexanoic acid tin (Japanese east transformation make, U-28) (divalent) 3 weight section and amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section was added, and the recovery was measured to it.

[0369] The constituent of the reference example 23 showed the recovery higher than the comparative example 17.

[0370] (The reference example 24 and the comparative example 18)

Organic polymer (A-10) 95 weight section which has the reactive silicon group obtained in the synthetic example 10, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, Hakukenka COR) 80 weight section, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, BUSUKO light R) 60 weight section, Heavy-calcium-carbonate (product made from Shiraishi industry, HOWATON SD) 20 weight section, DOP-40 weight section, epoxy system plasticizer (New Japan Chemical make, SANSON sizer EP-S) 20 weight section, thixotropic grant agent (made in [Kusumoto Chemicals] 1, DISUPARON 305) 3 weight section, and a photo-setting resin (the Toagosei make), ARONIKUSU M-3093 weight section, light stabilizer (Sanjyo make, SANORULST701) 1 weight section, Ultraviolet ray absorbent (made in [The Specialty Chemicals] 1, thavin 327) 1 weight section and antioxidant (made in [The Specialty Chemicals] 1, IRUGA NOX 1010) 1 weight section were measured, respectively, and it then kneaded with a 3 point roll, and was considered as base resin.

[0371] 2-ethylhexanoic acid tin (Japanese east transformation make, U-28) (divalent) 3 weight section and amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section and thixotropic grant agent (made in [Kusumoto Chemicals] 1, DISUPARON 305) 3 weight section, and a photo-setting resin (the Toagosei make), ARONIKUSU M-3093 weight section, light stabilizer (Sanjyo make, SANORULST701) 1 weight section as a hardening agent is made into the reference example 24, the thing using the mixture of 2-ethylhexanoic acid tin (Japanese east transformation make, U-28) (divalent) 3 weight section and amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section as a hardening agent was made into the comparative example 18. Base resin and a hardening agent were mixed uniformly and the recovery and then layer hardenability were evaluated.

[0372] The constituent of the reference example 24 showed good thin layer hardenability rather than the comparative example 18, while the high recovery was shown.

[0373] (Synthetic example 13)

To the allyl end and polyisobutylene obtained according to the example of manufacture of JP-H1-200639A, under existence of Pt catalyst, it was made to react to triethoxysilane and the polyisobutylene (A-13) which has a triethoxy allyl group at the end was obtained.

[0374] (Synthetic example 14)

To the allyl end and polyisobutylene obtained in the synthetic example 13, under existence of Pt catalyst, it was made to react to methyl dimethoxysilane and the polyisobutylene (A-14) which has a methyl dimethoxy allyl group at the end was obtained.

[0375] (Reference example 25 and the comparative example 19)

To organic polymer (A-13, A-14) 100 weight section which has the reactive silicon group obtained in the synthetic example 13 and the synthetic example 14, the amount part of divinylbenzene-bisacrylate (Japanese east transformation make, neo SUTAN U-220) duplex was added, and the hardened material was obtained. The thing using (A-13) as an organic polymer was made into the reference example 25, and the thing using (A-14) was made into the comparative example 18. The hardened material of the reference example 25 showed the recovery higher than the comparative example 19.

[0376] (Synthetic example 15)



